Mid-IR lasers: Challenges Imposed by the Population Dynamics of the Gain System

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Report Documentation Page

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Outline

1. Introduction: Level scheme, spectroscopic processes
2. Erbium 3-µm fiber lasers: Depleting the lower laser level
3. The cascade-lasing regime
4. The lifetime-quenching regime
5. The energy-recycling regime
6. The thermal problem
7. Another cascade-lasing regime
8. Other 3-µm fiber laser
9. Conclusions
## Lanthanide Ions in the Periodic System

Lanthanide $= 4f$ rare-earth ion, $^{57+n} \text{Ln} \ [_{54}\text{Xe} \ 6s^2 \ 5d \ 4f^n \ or \ _{54}\text{Xe} \ 6s^2 \ 4f^{n+1}]$
1. Shielding vs. core electric charge by inner shells (1s, ..., 4d)

⇒ central-field approximation
   (neglects perturbations within 4f sub-shell)

2. Shielding vs. interactions with host lattice
   by outer 5s and 5p sub-shells

⇒ relatively small electron-phonon coupling
The energy of the 4f subshell can be calculated in the central field approximation.

This approach takes into account the electric field produced by the charge of the nucleus and the charges of the filled inner shells which shield the charge of the nucleus.

It neglects the coulomb interaction between electrons within the partially filled 4f subshell, their spin-orbit coupling, and the crystal field generated by the ligand ions in the host material.
1. a) Non-centrosymmetric splitting (Coulomb interaction) ⇒ total orbital angular momentum
   \[ \mathbf{L} = \sum \mathbf{\ell} \]
   b) Accordingly: ⇒ total electron-spin momentum
   \[ \mathbf{S} = \sum \mathbf{s} \]

2. Spin-orbit coupling ("LS" coupling) ⇒ total angular momentum
   \[ \mathbf{J} = \mathbf{L} + \mathbf{S} \]
   lanthanides: intermediate coupling \((LS / jj)\)

3. Crystal-field splitting ("Stark effect") ⇒ total magnetic dipole moment
   \[ \mathbf{m}_J \]
Amounts of Splittings

1. Non-centrosymmetric splitting                ~ 10000 cm$^{-1}$
2. Spin-orbit splitting                                      ~ 1000 cm$^{-1}$
3. Crystal-field splitting                                    ~ 100 cm$^{-1}$

Unit (cm$^{-1}$) : photon energy $E = \frac{hc}{\lambda} \propto \frac{1}{\lambda}$

5000 cm$^{-1} = 2$ μm
10000 cm$^{-1} = 1$ μm
20000 cm$^{-1} = 500$ nm
Partial Energy-Level Scheme of Er$^{3+}$
Boltzmann Factors

At room temperature (300 K), \( k_B T = 200 \text{ cm}^{-1} \) is in the order of the energy splitting within a crystal-field multiplet

⇒ Boltzmann distribution of excitation energy within each crystal-field multiplet \( ^{2S+1}L_J \)

\[
f_i = \frac{\exp\left[\frac{(E_1 - E_i)}{(k_B T)}\right]}{\sum_i \exp\left[\frac{(E_1 - E_i)}{(k_B T)}\right]}
\]
Population Mechanisms

Stimulated processes:
- Ground-state absorption
- Excited-state absorption
- Stimulated emission

Spontaneous processes:
- Luminescence decay
- Multiphonon relaxation

Interionic processes:
- Energy migration
- Cross-relaxation
- Energy-transfer upconversion
Luminescence Decay

Einstein coeff. $A$ for spontaneous emission

$$\sum_{j} A_{ij} = A_i = \tau_{i,\text{rad}}^{-1}$$

radiative rate constant

Lifetime

$$\tau_{i}^{-1} = A_i + W_i$$  \hspace{1cm} \text{(rad. + nonrad. decay)}

Radiative rate constant, emission cross-section, oscillator strength are directly connected with each other.

Decay rate:

$$R_{ij} = -A_{ij}N_i$$
Multiphonon Relaxation: Consequences for Lifetime

Example:

Decay from the $^4I_{9/2}$ level of Er$^{3+}$
(to next lower-lying level: $\Delta E \approx 2000 \text{ cm}^{-1}$)

Competition between luminescence decay and multiphonon relaxation

Number of highest-energy phonons required to bridge $\Delta E$:

- Oxide: $p \approx 2 \Rightarrow \tau = 0.5 \mu s$
- Fluoride: $p \approx 4 \Rightarrow \tau = 7 \mu s$
- Chloride: $p \approx 7 \Rightarrow \tau = 4 \text{ ms}$
Upconversion Mechanisms

Intraionic process: ESA  
(Excited-state absorption)

Interionic process: ETU  
(Energy-transfer upconversion)
Energy-Transfer Processes

Interaction mechanisms:

1. multipole-multipole interaction:
   One oscillating multipole forces another nearby multipole to oscillate as well.

2. exchange interaction:
   Direct overlap between the atomic functions of two nearby ions

Most common:
Electric dipole-dipole interaction
Electric Dipole-Dipole Transfer

Transfer probability donor ($D$) $\rightarrow$ acceptor ($A$)

$$R_{DA} = \frac{3\hbar^4 c^4 Q_A}{4\pi n^4 \tau_D r_{DA}^6} \int f_D(E) F_A(E) E^{-4} dE$$


$Q_A = $ integral absorption cross-section of $A$
(measure of absorption probability)

$\tau_D = $ radiative lifetime of $D$
(measure of emission probability)

$f_D, F_A = $ normalized emission, absorption line shapes of $D, A$
(integral is measure of spectral overlap)

$r_{DA} = $ distance between $D$ and $A$  ($R_{DA} \propto r^{-6}$ !!!)
Energy Migration

Donor Ion

Pump

Acceptor Ion

Acceptor Ion
Sensitization and Quenching

Sensitization

Quenching

Sensitizing Ion A

Laser Ion B

Laser Ion A

Quenching Ion B

Pump

Pump
Cross-Relaxation and Energy-Transfer Upconversion

Cross-Relaxation

Energy-Transfer Upconversion

Donor Ion

Acceptor Ion

Pump

0

1

2

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Integrated Optical MicroSystems (IOMS)
Laser Wavelengths for Micro-Surgery

"Bottleneck" owing to longer lower level lifetime ⇒

"self-terminating" transition in continuous-wave operation.

CW inversion due to
1. weak feeding of lower level (host materials with low maximum-phonon energy)
2. Stark splitting

Depletion of lower laser level is desired to overcome bottleneck
The Erbium 3-µm Laser

Important processes:

Pump GSA @ 800 nm or 980 nm

Pump ESA @ 800 nm or 980 nm

ETU involving two ions in lower (1) or upper (2) laser level

Cross Relaxation (CR)
Depletion of Lower Laser Level

ESA  Energy Transfer  ETU  Laser

Ion 1  Ion 1  Ion 2  Ion 2

GSA  Laser  GSA  Laser  GSA  Laser  GSA

3  2  3  2  3  2  3

0  1  0  1  0  1  0

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Integrated Optical MicroSystems (IOMS)
Core-pumped fiber with typically
  0.1 mol. % (1000 ppm molar) (1.6×10^{19} \text{ cm}^{-3})

Low dopant concentration in combination with high-intensity
  core pumping favors ground-state bleaching

ESA becomes stronger than GSA

ETU is not important because of large distance between ions
Excited-State Absorption

GSA = Ground-State Absorption

ESA = Excited-State Absorption

SE = Stimulated Emission

\[ \lambda_1 = \text{pump wavelength}\]

(pump wavelength)

\[ \lambda_2 = \text{probe wavelength}\]

(broadband lamp)

GSA = Ground-State Absorption

ESA = Excited-State Absorption

SE = Stimulated Emission

\[ \lambda_1 \]

\[ \lambda_2 \]
ESAS measurement

S. Zemon, SPIE Vol. 1373 (1990) 21
Double Lock-in Amplifier Technique

2. Lock-in:
Ref. 11 Hz, detects $\Delta I$

1. Lock-in:
Ref. 390 Hz, detects $I_u$ or $I_p$

$\Delta I$

$I_u$

$I_p$

Up intensity I

time →
Determination of ESA Cross-Sections

Transmitted probe-beam intensity:

unpumped:

\[ I_u = I_0 \exp\{-d \cdot N_e \cdot \sigma_{ESA}\} \]

pumped:

\[ I_p = I_0 \exp\{d\left[ \left( 1 - N_e \right) \cdot \sigma_{GSA} - \sum_i N_i \cdot \sigma_{ESA,i} + \sum_i N_i \cdot \sigma_{SE,i} \right]\} \]

Calculation:

\[
\frac{1}{N_e \cdot d} \ln\left( \frac{I_u}{I_p} \right) + \sigma_{GSA} = \sum_i \left[ \frac{N_i}{N_e} \left( \sigma_{ESA,i} - \sigma_{SE,i} \right) \right]
\]

\textit{M. Pollnau, Appl. Phys. A 54 (1992) 404}

Determination of ESA Cross-Sections

Measure GSA cross-section $\sigma_{GSA}$, probe-beam intensities $I_u$, $I_p$

Fit excitation density $N_e$ until measured bleaching is completely compensated by addition of GSA (at wavelengths where no ESA or SE occurs)

Obtain ESA cross-sections $\sigma_{ESA,i}$ times relative population densities $N_i/N_e$

Determine relative population densities $N_i/N_e$ and calculate ESA cross-sections $\sigma_{ESA,i}$

*M. Pollnau, Appl. Phys. A 54 (1992) 404*

ESA at 800 nm in ZBLAN:Er$^{3+}$

\[ \begin{array}{c}
7 = 4^4 F_{3/2} \\
6 = 4^4 F_{5/2} \\
5 = 2^2 H_{11/2} \\
4 = 4^4 F_{9/2} \\
3 = 4^4 I_{9/2} \\
2 = 4^4 I_{11/2} \\
1 = 4^4 I_{13/2} \\
0 = 4^4 I_{15/2} \\
\end{array} \]

\( \tau = 570 \mu s \)

\( \tau = 6.7 \text{ ms} \)

\( \tau = 8.9 \text{ ms} \)

pump (990nm) probe (800nm)

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Integrated Optical MicroSystems (IOMS)
ESA Cross-Sections Near 800 nm

ESA cross sections determined from pump- and probe-beam measurements

M. Pollnau,

Best pump wavelength at 792 nm:

Relatively strong GSA,

Strong ESA from $^4I_{13/2}$,

Weak ESA from $^4I_{11/2}$
The Cascade-Lasing Regime

Two-loop cascade laser:

**ESA depletes lower laser level of 2.7-µm laser**

\[ ^4S_{3/2} \rightarrow ^4I_{13/2} \] (bypasses upper and populates lower laser level) is suppressed

- \( \tau_1 = 9.0 \, \text{ms} \)
- \( \tau_2 = 6.9 \, \text{ms} \)
- \( \tau_5 = 580 \, \mu\text{ms} \)
**Performance Under Cascade Lasing**

Ti:sapphire core-pumped ZBLAN fiber laser at 2.7 μm:

Strong increase of slope eff. and output power at the onset of cascade-lasing:

Slope eff. 23%

Output power 150 mW

*M. Pollnau, Appl. Phys. Lett. 66 (1995) 3564*
Cladding-pumped fiber with typically 1 mol. % (10000 ppm molar) (1.6×10^{20} \text{ cm}^{-3}) co-doped with Pr^{3+}

Higher dopant concentration in combination with low-intensity cladding pumping and lifetime quenching by Pr^{3+} favors ET

Ground-state bleaching and ESA are not important because of higher dopant concentration and low pump intensity
Lifetimes of $^4I_{11/2}$ upper and $^4I_{13/2}$ lower laser levels vs. Pr$^{3+}$ concentration

Quenching of lower level lifetime much stronger (from 9 ms down to 20 µs), because corresponding absorption transition in Pr$^{3+}$ has high oscillator strength

*P.S. Golding,*  
The Lifetime-Quenching Regime

Simple four-level laser:

Energy transfer to Pr$^{3+}$ co-dopant depletes lower laser level efficiently

CW threshold condition removes energy from $^4I_{11/2}$ upper laser level

ESA is avoided

*M. Pollnau,*

*IEEE JQE 33 (1997) 1982*
Diode-pumped double-clad ZBLAN fiber laser at 2.7 µm:

Low-brightness pump is converted to single-mode output

Slope eff. 17%

Output power 1.7 W

ZBLAN Fiber Laser at High Dopant Concentration

Cladding-pumped fiber with up to 10 mol. % (100000 ppm molar) (1.6×10^{21} \text{ cm}^{-3})

High dopant concentration favors ETU

Ground-state bleaching and ESA are not important because of high dopant concentration and low pump intensity
Measurement of ETU

Q-SWITCHED Nd:YAG LASER

532 nm FILTER

KTP OPO

532 nm

KD^P

532 nm FILTER

PHOTODIODE

MONOCHROMATOR

200 µm PINHOLE

COLLECTION OPTICS

POWER METER

DSO

Integrated Optical MicroSystems (IOMS)
Measured Luminescence Decay

Normalized decay curves:

First temporal part:
- high excitation density,
- includes decay by ETU,
- non-exponential

Last temporal part:
- low excitation density,
- includes no ETU,
- exponential

\[ P.S. \, Golding, \]
\[ Phys. \, Rev. \, B \, 62 \, (2000) \, 856 \]
Luminescence Decay Curves

Rate-equation for decay:
\[
dN / dt = -\tau^{-1} N - 2WN^2
\]

Solution (Bernoulli-Eq.):
\[
N(t) = \frac{N_0 \exp(-t / \tau)}{1 + 2WN_0 \tau\left[1 - \exp(-t / \tau)\right]}
\]

Linearized solution:
\[
\left[\frac{N_0}{N(t)}\right]\exp(-t / \tau) - 1 = 2WN_0 \tau\left[1 - \exp(-t / \tau)\right]
\]

\[
Y = A \cdot X
\]

Determination of ETU Parameters

Linearized solution:

\[ \left( \frac{N_0}{N(t)} \right) \exp(-t/\tau) - 1 = 2WN_0\tau[1 - \exp(-t/\tau)] \]

Determine

t from exponential part,

N0 from excited volume and absorbed pump energy,

Fit straight lines ⇒ W

Advantage of Procedure

If equation were not linearized, onset of repopulation from higher levels would not be seen

⇒ complete set of rate equations from all levels must be solved

⇒ one has to know all parameters (further ETU parameters !!!)

ETU and CR Processes

Measured parameters:

**CR** from $^4S_{3/2}$ level

**ETU** from $^4I_{11/2}$ upper laser level

**ETU** from $^4I_{13/2}$ lower laser level
ETU Parameters

ETU parameters determined from fluorescence decay

P.S. Golding, 

Quantum efficiency:

\[ \eta_q = 2 - \frac{b_1^2}{b_2^2} \frac{W_{22}}{W_{11}} \]

Important: Ratio \( W_{11}/W_{22} \)

M. Pollnau, 
IEEE JQE 32 (1996) 657
**The Energy-Recycling Regime**

**ETU processes from** lower laser level ($^4I_{13/2}$) recycles energy to upper laser level ($^4I_{11/2}$)

⇒ quantum efficiency of 2

⇒ increase in slope efficiency by factor of 2

*M. Pollnau, IEEE J. Select. Topics Quantum Electron. 7 (2001) 30*
Comparison: Thulium 2-µm vs. Erbium 3-µm Lasers

In both cases: increase in slope efficiency by factor of 2
Experimental Results in Crystals

Various host materials
(YLF, YAG, YSGG, GGG, ...)

~1 W output power


~50% slope efficiency


~2-4 W output power

A.Y. Dergachev, CLEO Technical Digest (2000), 564
Expected Fiber Performance Under Energy Recycling

Diode-pumped double-clad ZBLAN fiber laser at 2.7 µm:

Rate-equation calculation:
ETU can be exploited

Prediction:
Slope eff. 50%
Output power >10 W

M. Pollnau,
IEEE JQE 38 (2002) 162
The Thermal Problem

Strong heat load owing to multiphonon relaxations following the ETU processes

⇒ strong temperature increase

⇒ strong thermal lensing
  (crystal laser: $2 \times$ that of 1-µm Nd3+ laser!)

⇒ rod fracture

⇒ fiber melting
Example: LiYF$_4$ : Er$^{3+}$

Lasing conditions

Non-lasing conditions

*M. Pollnau, IEEE J Q-E 39 (2003) 350*
ZBLAN Fiber Laser at Medium Dopant Concentration

Cladding-pumped fiber with typically 0.5 mol. % (5000 ppm molar) (8×10^{19} \text{ cm}^{-3})

Lower dopant concentration in combination with low-intensity cladding pumping and cascade lasing at 1.6 \text{ μm} diminishes other spectroscopic processes

ESA and ETU are not that important because of low excitation density, ET is impossible because of lack of Pr^{3+}
The laser at 2.8 \( \mu m \) starts lasing first. As soon as the laser transition at 1.6 \( \mu m \) reaches threshold,

\( ^4I_{11/2} \) is clamped to threshold inversion vs. \( ^4I_{13/2} \).

\( ^4I_{13/2} \) is clamped to threshold inversion vs. \( ^4I_{15/2} \).

⇒ All three levels remain at constant density, heat input is minimal, no parasitic processes become important.
Input-Output Curves

Experimental

Calculated

S.D. Jackson et al., submitted (2010)

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Integrated Optical MicroSystems (IOMS)
Concerning heat input, there’s plenty of room for higher power!

S.D. Jackson et al., submitted (2010)
Uncooled ZBLAN fiber doped with 6 mol% Er$^{3+}$

9 W output power with 21% slope efficiency.


Cooled ZBLAN fiber doped with 6 mol% Er$^{3+}$

24 W output power with 16% slope efficiency.


Uncooled ZBLAN fiber doped with 0.5 mol% Er$^{3+}$

8 W output power with 19% slope efficiency, cascade laser.

_**S.D. Jackson et al., submitted (2010)**_
Other 3-µm Fiber Lasers

T. Sumiyoshi,
“High-power continuous-wave 3- and 2-µm cascade Ho\textsuperscript{3+}:ZBLAN fiber laser and its medical applications”,

S.D. Jackson,
“Single-transverse-mode 2.5 W holmium-doped fluoride fiber laser operating at 2.86 µm”,

S.D. Jackson,
“Continuous wave 2.9 µm dysprosium-doped fluoride fiber laser”,
Summary

Population mechanisms of Er$^{3+}$ 3-µm fiber laser depend on Er$^{3+}$ conc., pump parameters, fiber geometry

⇒ we need to understand its spectroscopy!

With increasing erbium conc., four different operation regimes have successfully been demonstrated experimentally:

1. core pump, ESA ⇒ upper cascade lasing
2. clad pump, codoping ⇒ lifetime quenching
3. clad pump, ETU ⇒ energy recycling
4. clad pump, laser depletion ⇒ lower cascade lasing

Current output power is at 9 W uncooled and 24 W cooled.